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THEORY OF DIELECTRIC BREAKDOWN IN REACTIVE MEDIA.

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BY **MICHAEL J. FRANKEL**

RESEARCH AND TECHNOLOGY DEPARTMENT

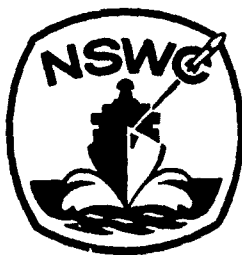
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solutions to the heat transport equation are obtained and analyzed.

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SUMMARY

We have examined the theory of both electrical and thermal breakdown in explosive materials. Application of thermal breakdown theory had to be modified to account for the reaction kinetics while electrical breakdown, whose onset occurs at the steady state temperature, was found to follow breakdown theory in inerts.

Use of breakdown theory to predict critical fields in explosives seems feasible but is hampered by the lack of an adequate data base. This lack of experimental data also prevents a resolution of the question of possible electric field effects on thermal explosion parameters. It should, however, be relatively simple to distinguish between thermal and electrical breakdown in any given experiment by a consideration of initial temperature and explosion induction time.

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I. INTRODUCTION

Dielectric breakdown is a universal response of materials to the application of sufficiently intense electromagnetic fields. While the interaction of electromagnetic radiation with inert dielectrics has received attention from both theorist¹⁻⁶ and experimentalist⁷⁻⁹, relatively little work has been done on the coupling of radiation to explosive materials, a matter of some recent interest. We may also note experimental evidence indicating generation of intense polarization fields in a detonating explosive¹⁰⁻¹². An understanding of breakdown phenomena may thus help elucidate fundamental detonation processes, even in the absence of externally coupled fields.

Theories of breakdown in solids divide broadly into studies of thermal breakdown and purely electrical breakdown, with a number of subcategories for the latter. We shall briefly review each process and consider the application to an explosive medium.

II. ELECTRICAL BREAKDOWN

1. FORMULATION OF BREAKDOWN CRITERION. In electrical breakdown we assume that the electron distribution depends directly on the applied electric field \vec{E} . The critical breakdown

¹Frohlich, H., "Theory of Electrical Breakdown in Ionic Crystals," Proc. Roy. Soc. A160, 230 (1937).

²Frohlich, H. and Paranjape, V., "Dielectric Breakdown in Solids," Proc. Phys. Soc. Lond. B69, 866 (1956).

³O'Dwyer, J. J., "Dielectric Breakdown in Solids," Adv. in Phys. 7, 349 (1958).

⁴Seitz, F., "On the Theory of Election Multiplication in Crystals," Phys. Rev. 76, 1376 (1949).

⁵Stratton, J. A., "Theory of Dielectric Breakdown in Solids," Progress in Dielectrics, Vol. 3, Ed. Birks (1961, Wiley).

⁶Von Hippel, A., "The Electrical Breakdown Strengths of Ionic Crystals as Functions of Temperature," Phys. Rev. 56, 941 (1939).

⁷Vorobev, A., "Anisotropy of the Dielectric Strength of Rocksalt," Soviet Phys. Solid State 4, 1441 (1962).

⁸Caspari, M., "Direction of Breakdown in Alkali Halide Crystals," Phys. Rev. 98, 1679 (1955).

⁹Davisson, J. W., "Directional Breakdown in Crystals," Progress in Dielectrics Vol. 1, Ed.-Birks, (1959-Wiely).

¹⁰Hayes, B., "The Detonation Electric Effect," J. App. Phys. 38, 507 (1967).

¹¹Dremin, A. et.al, "Shock Induced Electrical Polarization," Proc. 6th Int. Symp. on Detonation, (1975).

¹²Yakushev, V., et.al, "On the Measurement of Polarization Relaxation Time in Homogeneous Explosives," Zh. Experm. Teor. Fiz. 54, 396 (1961).

field \vec{F}_0 will be that marking the onset of some instability in the conduction current.

We consider first the case of low carrier density and follow the average behavior of a single electron interacting with the applied electric field and with the lattice. As the density increases, electron-electron interactions become important and a single electron picture will no longer suffice. We will later calculate the critical density for a single electron approximation and consider the necessary modifications when the low density restriction is violated.

For a steady state to exist, an equilibrium must be maintained between the energy supplied to the electron by the external field $A(\vec{F}, E)$, and the net energy transferred from the electron to the lattice via collisions with phonons, which we shall designate $B(E)$. E represents the total electronic energy.

We consider an applied electric field \vec{F} pointing along the z-axis of our cartesian coordinate system. A conduction band electron will be accelerated by the field feeling a force

$$\frac{dp_z}{dt} \cdot \text{FIELD} = eF \quad (1)$$

We adopt the convention that a previously defined vector symbol appearing without an arrow refers to the absolute value. The buildup of momentum in the z-direction will be retarded through collision with the lattice leading to an average drag force

$$\frac{dp_z}{dt} \text{ LATTICE} = \frac{-p_z}{\tau(E)} \quad (2)$$

where we have defined an average collision time $\tau(E)$ which is a function of the electronic energy E .

Equilibrium requires

$$\left(\frac{dp_z}{dt} \right) \text{ LATTICE} + \left(\frac{dp_z}{dt} \right) \text{ FIELD} = 0 \quad (3)$$

We define a current density $\vec{J}(E)$ by

$$\vec{J}(E) = \frac{e\vec{p}}{m} \quad (4)$$

where m is the effective mass of an electron in a conduction band. The energy given up by the electric field may be obtained in a straightforward manner from the Maxwell equations as $\vec{J} \cdot \vec{E}$. Using Equations (2)-(4) we then have for the net energy per unit volume transferred from the field to the electron

$$A(\vec{F}, E) = \frac{e^2 F_T(E)}{m} \quad (5)$$

The net rate of energy transfer to the lattice may be calculated by summing over all lattice modes, the probability that the electron will either absorb or emit a phonon of wave vector \vec{q} .

$$B(E) = \sum_{\vec{q}} \hbar \omega_{\vec{q}} P_{(k-\vec{q}, k)}^{\text{emission}} - P_{(k+\vec{q}, k)}^{\text{absorption}} \quad (6)$$

The total Hamiltonian of the system is taken to be

$$H = H_{\text{electron}} + H_{\text{phonon}} + H_{\text{e-ph}} \quad (7)$$

with $H_{\text{e-ph}}$ representing the electron-phonon interaction, considered small relative to the total Hamiltonian.

The quantum mechanical probabilities of emission and absorption are then given by the usual "Golden Rule" transition rate formula

$$P^e(k-\vec{q}, k) = \frac{2\pi}{\hbar} |\langle k-\vec{q} | H_{\text{e-ph}} | k \rangle|^2 (n_{\vec{q}} + 1) \times \delta\left\{\frac{\hbar^2}{2m}(\vec{k}-\vec{q})^2 + \hbar\omega_{\vec{q}} - \frac{\hbar^2 k^2}{2m}\right\} \quad (8)$$

$$P^a(k+\vec{q}, k) = \frac{2\pi}{\hbar} |\langle k+\vec{q} | H_{\text{e-ph}} | k \rangle|^2 n_{\vec{q}} \delta\left\{\frac{\hbar^2}{2m}(\vec{k}+\vec{q})^2 - \hbar\omega_{\vec{q}} - \frac{\hbar^2 k^2}{2m}\right\} \quad (9)$$

The matrix element $\langle \vec{k} \pm \vec{q} | H_{\text{e-ph}} | \vec{k} \rangle$ represents the probability amplitude for an electron initially in a state of wave vector $|\vec{k}\rangle$ to scatter to a final state of wave vector $|\vec{k} \pm \vec{q}\rangle$. δ is the Dirac delta function ensuring conservation of energy and $n_{\vec{q}}$ is a phonon density function given by the quantum statistical distribution for Bose particles

$$n_{\vec{q}} = \frac{1}{e^{\hbar\omega_{\vec{q}}/kT} - 1} \quad (10)$$

The condition for the existence of a steady state is then given by

$$\frac{e^2 F^2 \tau(E)}{m} = \frac{2\pi}{\hbar} \sum_q \hbar \omega_q \{ |\langle k-q | H_{e-ph} | k \rangle|^2 (n_q + 1) \delta_1 - |\langle k+q | H_{e-ph} | k \rangle|^2 n_q \delta_2 \} \quad (11)$$

δ_1 and δ_2 refer to the delta functions of Equation (8) and (9) respectively. To complete the calculation it is necessary to specify functional forms for $J(E)$ and for the interaction matrix elements.

The left side of Equation (2) represents the rate of change of momentum p_z due to electron-phonon interactions. The total rate of electronic momentum change may be obtained by summing the probabilities that the electron will either absorb or emit any lattice phonon. We may thus rewrite Equation (2) in the form

$$\begin{aligned} \frac{1}{\tau(E)} = \frac{2\pi}{\hbar} \sum & \frac{(k_f - k)}{k} |\langle k-q | H_{e-ph} | k \rangle|^2 (n_q + 1) \delta_1 \\ & + \frac{(k_f + k)}{k} |\langle k+q | H_{e-ph} | k \rangle|^2 n_q \delta_2 \quad (12) \end{aligned}$$

2. THE ELECTRON-PHONON INTERACTION.

a. Polar Molecules. The electron-phonon interaction term will depend on the polarization properties of the molecular lattice. In polar molecules the longitudinal optical modes produce long range dipole fields with which the electron may be strongly coupled, and interaction with acoustic phonons is neglected.

The energy of an electron in a field is given by the expression

$$H_{e-ph} = e\phi(r) \quad (13)$$

where $\phi(r)$ is the scalar potential function. The divergence \vec{D} is related to the polarization field \vec{P} by

$$\vec{D} = \vec{E} + 4\pi\vec{P} \quad (14)$$

The vanishing of the divergence then gives the polarization produced by the optical phonons as

$$\vec{P} = \frac{1}{4\pi} \vec{\nabla} \phi \quad (15)$$

The polarization and the potential are expanded in fourier components

$$\vec{P}(\vec{r}) = \frac{C}{\sqrt{V}} \sum_{\vec{q}} \hat{e}_{\vec{q}} (a_{\vec{q}} e^{i\vec{q} \cdot \vec{r}} + a_{\vec{q}}^+ e^{-i\vec{q} \cdot \vec{r}}) \quad (16)$$

$$\phi(r) = \frac{1}{\sqrt{V}} \sum_{\vec{q}} \phi_{\vec{q}} e^{i\vec{q} \cdot \vec{r}} + \phi_{\vec{q}}^+ e^{-i\vec{q} \cdot \vec{r}} \quad (17)$$

C is a constant which will be determined and the transformed coordinates $a_{\vec{q}}, a_{\vec{q}}^+$ correspond to the phonon annihilation and creation operators in second quantized representation. Substituting (14) and (15) into (13) we obtain

$$\phi_{\vec{q}} = \frac{-4\pi i C}{q} a_{\vec{q}}, \phi_{\vec{q}}^+ = \frac{4\pi i C}{q} a_{\vec{q}}^+ \quad (18)$$

Substituting (15) and (16) into (11) gives

$$H_{e-ph} = \frac{-1}{\sqrt{V}} \sum_{\vec{q}} \frac{4\pi i e C}{q} \{ a_{\vec{q}} e^{i\vec{q} \cdot \vec{r}} - a_{\vec{q}}^+ e^{-i\vec{q} \cdot \vec{r}} \} \quad (19)$$

The constant C is calculated in Appendix A and is shown to be

$$C = \frac{1}{4\pi} \left(\frac{\hbar\omega}{2} \right)^{\frac{1}{2}} \left(\frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon} \right)^{\frac{1}{2}} . \quad (20)$$

Equation (19) gives the Frohlich interaction term¹³ for polar crystals.

Using (17) and the selection rules for harmonic oscillator transitions (cf. Appendix A) we have for the absorption and emission transition rates

$$P^a = \frac{2\pi}{\hbar} \frac{e^2}{V} \frac{\hbar\omega}{2} \left[\frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon} \right] \frac{n_q \delta_1}{q^2} , \quad (21)$$

$$P^e = \frac{2\pi}{\hbar} \frac{e^2}{V} \frac{\hbar\omega}{2} \left[\frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon} \right] \frac{(n_q + 1)}{q^2} \delta_2 . \quad (22)$$

b. Non-Polar Molecules. F. Seitz has shown⁵ that H_{e-ph} will arise largely from the electron-acoustic phonon coupling in non-polar molecules. Let $V(\vec{r} - \vec{x}_{\mu,l})$ equal the potential energy of an electron at position \vec{r} due to the μ th ion in the l th unit cell. $\vec{x}_{\mu,l}$ denotes the lattice vector to the origin of the unit cell.

Let

$$\vec{x}_{\mu,l} = \vec{R}_{\mu,l} + \vec{u}_{\mu,l} , \quad (23)$$

where $\vec{u}_{\mu,l}$ represents the displacement from the equilibrium position \vec{R}_{μ} . To first order the total potential at \vec{r} may then be given by

¹³Frohlich, H., "Electrons in Metals," Adv. in Phys. 3,325 (1954).

$$V(r) = \sum_{\ell, \mu} V(r - X_{\mu, \ell}) = \sum_{\ell, \mu} V(r - R_{\mu, \ell}) + \vec{u}_{\mu, \ell} \cdot \vec{\nabla} V(r - R_{\mu, \ell}) . \quad (24)$$

We see that the lattice vibrations induce an energy shift from the value perceived by an electron in the undistorted lattice. This energy shift

$$\Delta V = \sum_{\ell, \mu} \vec{u}_{\mu, \ell} \cdot \vec{\nabla} V(r - R_{\mu, \ell}) \quad (25)$$

is considered to be the quantum mechanical perturbation operator, where the displacement $\vec{u}_{\mu, \ell}$ is a phonon operator and $\vec{\nabla} V$ operates on the electronic states only.

The phonon operator $\vec{u}_{\mu, \ell}$ is expanded in normal coordinates giving

$$\Delta V = \sum_{\ell, \mu} \vec{\nabla} V(r - R_{\mu, \ell}) \sum_{\alpha} \left(\frac{\hbar}{2MN\omega_{\alpha}} \right)^{1/2} e^{iq \cdot R_{\mu, \ell}} \epsilon_{\alpha} (a_{\alpha}^{\dagger} - a_{\alpha}) ; \quad (26)$$

α represents the sum over both wave-vector and polarization and M is the ionic mass. When (26) is substituted into the matrix element for electron-phonon coupling one obtains for the transition probabilities

$$p^e = \frac{2\pi}{\hbar} \frac{I^2 \hbar q (n_q + 1) \delta^1}{2M s v n} , \quad (27)$$

$$p^a = \frac{2\pi}{\hbar} \frac{I^2 \hbar q n_q \delta^2}{2M s v n} . \quad (28)$$

In deriving (27), (28) we used the acoustic-phonon dispersion relation $\omega_q = sq$ with s the sound velocity. Use of¹⁴ Green's Theorem allows conversion of the electron state matrix element involving $\vec{\nabla} V$ into an electron overlap integral of the form

$$I = D \int \psi_k^* \psi_{k'} d^3r . \quad (29)$$

¹⁴Ziman, J. M., Electrons and Phonons, Oxford V. Pres., (1960).

Estimates of I in semiconductors¹⁵ give values of the order of one electron volt.

3. RELAXATION TIMES. The term $(k_f - k)/k$ appearing in the expression for the relaxation time (Equation (12)) may be obtained from energy and momentum conservation. Conservation of energy gives

$$\cos(k, q) = \frac{-q}{2K} \pm \frac{m\hbar\omega}{kq}, \quad (30)$$

where the minus or plus sign refers to emission or absorption, respectively. Averaging over the azimuth⁶ angle gives

$$\frac{k_z}{k} = \frac{-q^2}{2k^2} \mp \frac{m\hbar\omega}{k^2}. \quad (31)$$

We assume a dense distribution of modes q and pass from a sum to an integral with the standard approximation

$$\begin{aligned} \sum_q &\rightarrow \frac{V}{(2\pi\hbar)^3} \int d^3p \\ &= \frac{V}{(2\pi\hbar)^3} \pi \int p^2 dp d(\cos \theta), \end{aligned} \quad (32)$$

where $p = \hbar q$ is the phonon momentum and θ is the angle between k and q , the initial electron momentum and phonon wave vector. The integrations over the probability functions, either of the form of (21-22) or (27-28), involves an integral over a delta function, $\delta(\frac{\hbar^2}{2m}(\vec{k} \pm \vec{q})^2 - \frac{\hbar^2 k^2}{2m} \pm \hbar\omega_q)$, as defined in (8-9). The integral over $\cos \theta$ can then be written

¹⁵ Ershov, A. P., "Ionization During Detonation of Solid Explosives," *Fixiba Gor. Vary*, 11, 938 (1974).

$$\begin{aligned}
& \int d(\cos \theta) \delta \left\{ \frac{\hbar^2 q^2}{2m} \pm \hbar \omega_q \pm \frac{\hbar^2 k q}{m} \cos \theta \right\} \\
&= \int \frac{d(\cos \theta) \delta \left\{ \cos \theta \mp \frac{\hbar \omega_q m}{\hbar^2 k q} + \frac{\hbar^2 q^2 m}{2m \hbar^2 h q} \right\}}{\hbar^2 k q / m} ,
\end{aligned} \tag{33}$$

where we have used the well known property of the delta function

$$\delta\{f(x)\} = \sum_i \frac{\delta(x - x_i)}{|f'(x_i)|} , \tag{34}$$

where x_i are the roots of $f(x)$.

The integration over $\cos \theta$ then gives

$$\int d(\cos \theta) \delta(f(\theta)) = \frac{m}{\hbar^2 k q} . \tag{35}$$

Equation (12) may now be written

$$\begin{aligned}
\frac{1}{\tau(E)} &= \frac{V m}{2(2\pi)^2 \hbar^2 k^3} \left\{ \int q^3 G_q \left(1 + \frac{2m\omega}{\hbar q^2}\right) (n_q + 1) dq \right. \\
&\quad \left. + \int q^3 G_q \left(1 - \frac{2m\omega}{\hbar q^2}\right) n_q dq \right\} ,
\end{aligned} \tag{36}$$

where G_q is just the probability function without the delta function factor.

For electron energies $E \gg \hbar \omega_q$ the factors $1 \pm 2m\omega/hq^2$ will both be approximately equal to one. The integral may then be approximated by

$$\frac{1}{\tau(E)} \approx \frac{V_m}{2(2\pi)^2 \hbar^2 k^3} \int q^3 G_q (2n_q + 1) dq . \quad (37)$$

For long wave length phonons the integrand goes to zero. We may therefore set the lower limit of integration equal to zero while the upper limit may be approximated by

$$q_{\max} \approx 2k . \quad (38)$$

For polar molecules it is found that the optical phonons are relatively dispersionless. The factor $2n_q+1$ may then be removed from the integral. Using (21-22) in (36) we have

$$\frac{1}{\tau(E)} = \frac{\alpha}{E^{\frac{1}{2}}} \left\{ 1 + \frac{2}{e^{\hbar\omega/kT} - 1} \right\} , \quad (39)$$

where

$$\alpha = \frac{e^2 \sqrt{m\omega}}{4\pi\sqrt{2}} \left(\frac{1}{\epsilon^\infty} - \frac{1}{\epsilon} \right) . \quad (40)$$

For non polar molecules we use Equation (27-28) and approximate

$$n_q = \frac{1}{e^{\hbar sq/kT} - 1} \approx \frac{kT}{\hbar sq} . \quad (41)$$

The relaxation integral then takes the form

$$\frac{1}{\tau(E)} = \frac{\sqrt{2}(m)^{3/2} I^2 k_B T \sqrt{E}}{2\pi S^2 N m \hbar^4} , \quad (42)$$

where it has been assumed that the main acoustical interaction is with the long wavelength phonons.

4. RATE OF ENERGY TRANSFER TO THE LATTICE. The energy transfer rate to the lattice may now be written down as

$$B(E) = \frac{V_m}{(2\pi)^2 \hbar^2 k} \int q G_q \hbar \omega dq . \quad (43)$$

In polar crystals the integral diverges as $q \rightarrow 0$. q_{\min} is then obtained by conservation of energy (equation along with the approximations)

$$k + k' = 2k , \quad (44)$$

$$k - k' = q_{\min} . \quad (45)$$

This gives as the lower limit in (40)

$$q_{\min} = \frac{m\omega}{\hbar k} . \quad (46)$$

Performing the integration we then have the following expressions for $B(E)$. In polar crystals

$$B(E) = \frac{\sqrt{m} e^2 \omega^2}{4\pi \sqrt{2}} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon} \right) \frac{1}{\sqrt{E}} \log \frac{4E}{\hbar \omega} . \quad (47)$$

In non-polar crystals, taking the lower limit of the integral equal to zero we have

$$B(E) = \frac{I^2 (2m)^{3/2} m E^{3/2}}{\pi \hbar^4 N M} .$$

5. CALCULATION OF CRITICAL FIELDS. The critical field F_B for breakdown is now calculated from the energy balance Equation (11) with a particular choice of electron energy E . If E is chosen equal to the ionization energy E_I we obtain the following expressions for the critical field dependence on energy and temperature. For polar modes:

$$F_C = \frac{me\omega^{3/2}}{4\pi\sqrt{2}\hbar} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon} \right) \frac{1}{\sqrt{E_I}} \left\{ \log \frac{4E_I}{\hbar\omega} \right\}^{1/2} \left\{ 1 + \frac{2}{e\hbar\omega/kT-1} \right\}^{1/2} \quad (48)$$

For acoustic modes:

$$F_C = \frac{m^{3/2}I^2(k_B T)^{1/2}}{2^{1/4}\pi e\hbar^4 s} \frac{m}{M} E_I \quad (49)$$

In Figures 1 and 2, we have graphed the behavior of the critical field as a function of temperature for a reasonable choice of physical parameters.

To illustrate the use of Equation (48) for a typical polar molecule, we will estimate the breakdown voltage for TNT. In Figure 1, we have chosen E_I as 6e.v.¹⁵ and have taken for ω a typical frequency of 10^{14} . The other parameters chosen were $T=300^\circ\text{K}$, $m=m$ (electron) and $(1/\epsilon_\infty - 1/\epsilon) \approx \frac{1}{2\epsilon_0}$ where ϵ_0 is the free-space permittivity. We then obtain for the critical breakdown field, $F_C \approx 10^6$ V/cm. This value should be accurate to within an order of magnitude given the uncertainty in the physical parameters.

6. TIME LAG BEFORE BREAKDOWN. We shall make a rough estimate of the time till breakdown assuming a critical field $F_B \approx 10^6$ v/cm has been applied.

Let the electron be accelerated from rest and assume that it ionizes another electron immediately upon attainment of the critical energy

$$\frac{1}{2}mV_B^2 = I \quad (50)$$

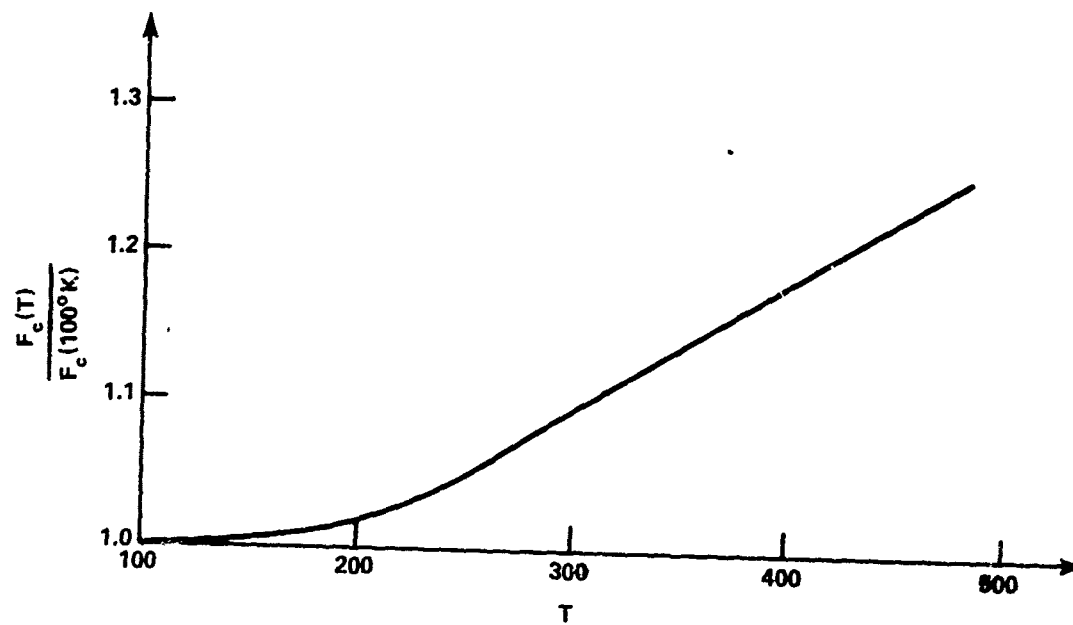


FIGURE 1 CRITICAL BREAKDOWN FIELD AS FUNCTION OF TEMPERATURE (°K) IN POLAR CRYSTALS. $F_c(100^\circ K)=10^6 V/cm$

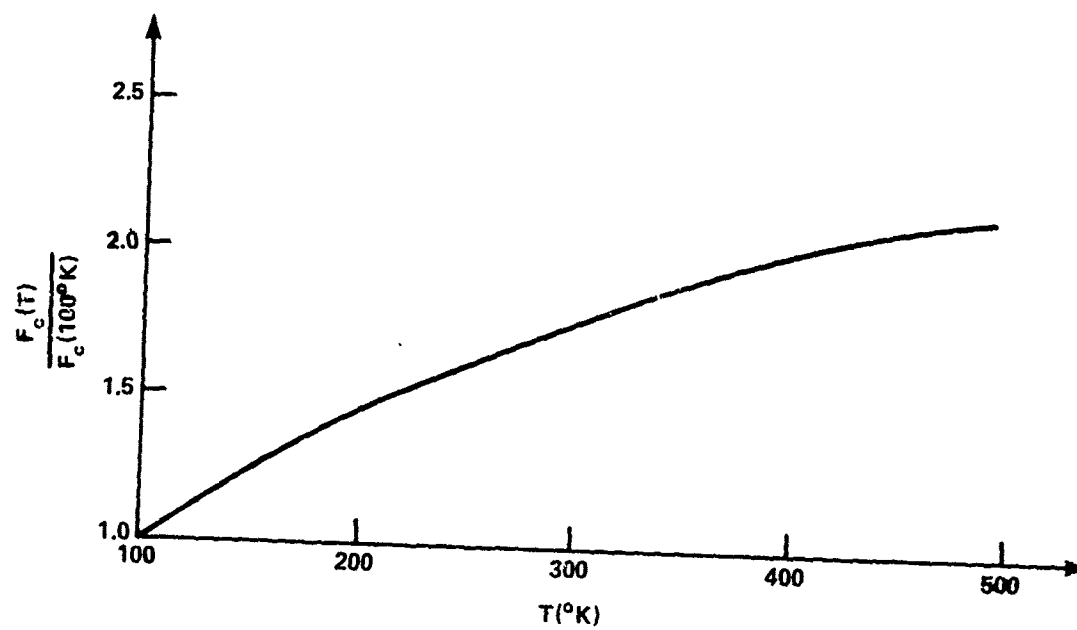


FIGURE 2 CRITICAL BREAKDOWN FIELD AS FUNCTION OF TEMPERATURE ($^{\circ}K$) IN NON-POLAR CRYSTALS. $F_c(100^{\circ}K) = 0.6 \times 10^6 V/cm$

If we take $I = 6\text{e.v.}$ characteristic of TNT then

$$V_B = \sqrt{\frac{2I}{m}} \approx 1.4 \times 10^6 \text{ m/s} . \quad (51)$$

With a relaxation time $\tau = 10^{-14}$ sec, an electron will achieve a velocity

$$V_B = \frac{eF_B}{m} \tau = 1.7 \times 10^5 \text{ m/s} \quad (52)$$

before collision, which is not enough to achieve ionization. We must then ask how many collisions, on the average, will occur before an electron will have sufficient time t_B between collisions to achieve the critical velocity V_B . We can estimate this by writing down a probability function $P(t)$ representing the probability that a time t has passed with no collisions. We shall take a model $P(t)$ as

$$P(t) = e^{-t/\tau} , \quad (53)$$

which has the proper boundary behavior

$$P(0) = 1 \quad \text{and} \quad P(\infty) = 0 , \quad (54)$$

and which also falls off rapidly for $t > \tau$. We may also formally derive Equation (53) by noting that $P(t+dt)$ represents the probability that no collision will occur in a time t or in a following time interval dt . We thus have

$$P(t+dt) = P(t) P(dt) . \quad (55)$$

expanding $P(dt)$ about $t = 0$ and applying the boundary condition we have

$$P(t+dt) = P(t) \left\{ 1 + \frac{dP}{dt} \bigg|_{t=0} dt \right\} . \quad (56)$$

If we make the identity $\tau^{-1} = \frac{dP}{dt} \bigg|_{t=0}$ we may formally obtain the equation

$$\frac{dP}{dt} = \frac{-P(t)}{\tau} . \quad (57)$$

Integrating (57) gives us the probability function of Equation (53).

The time needed for a single electron to achieve breakdown is given by

$$t_B = mV_B/eF_B \approx 9 \times 10^{-14} \text{ sec} \quad (58)$$

with the values of V_B and F_B given above. We thus have

$$P(t_B) = e^{-9} \approx 1/8000 . \quad (59)$$

It will take on the average 8000 collisions to produce a single ionization event. The time for each ionization event is thus given by

$$t_I = 8000\tau = 8 \times 10^{-11} \text{ sec} . \quad (60)$$

To estimate now the total number of ionizing collisions n which will characterize dielectric breakdown of the material we equate the energy supplied by the field to the electrons with the energy required to raise the lattice to some critical temperature T_0 . After n collisions we have 2^n free electrons so we set

$$2^n I = c_v \rho V (T_0 - T) . \quad (61)$$

V represents the volume of the cone to which the 2^n electrons are assumed confined. The breakdown time τ_B will be given by

$$\tau_B = nt_I . \quad (62)$$

The volume of the cone V is given by

$$V = 1/3\pi (\bar{v}\tau_B)^3 \tan^2 \alpha \quad (63)$$

where \bar{v} is the average velocity in the field direction given by the condition

$$\bar{v} = \frac{1}{2} v_B \quad (64)$$

and α is the half angle of the cone. $\tan \alpha$ may be evaluated by assuming that motion perpendicular to the applied field is governed only by thermal diffusion. The thermal velocity v_t is determined by

$$v_t = \left(\frac{2kT}{m} \right)^{\frac{1}{2}} \quad (65)$$

After a time τ_B corresponding to n collisions the electrons may diffuse out to a distance

$$l = n^{\frac{1}{2}} \frac{2kT}{m}^{\frac{1}{2}} \tau_B \quad (66)$$

where the $n^{\frac{1}{2}}$ term comes from the drunkard's walk solution. We then have

$$\tan \alpha = (2knT)^{\frac{1}{2}} / \frac{1}{2} v_B \quad (67)$$

Substituting (62-67) into (61) and taking the log of both sides, we obtain a transcendental equation for n

$$n - \frac{4 \log n}{\log 2} = \log \left\{ \frac{c_v \rho \Delta T \pi k_B T}{3Im} v_B \tau_B^3 \right\} / \log 2 \quad (68)$$

We take $c_v = .2$ cal/gm, $\rho = 1.6$ gm/cm³, $\Delta T = 100^\circ\text{K}$ with the other constants as before. We have solved Equation (8) graphically (see Figure 3) and obtain for n

$$n \approx 52 \quad (69)$$

We then have

$$\tau_B \approx 52 \tau_B \approx 4 \times 10^{-9} \text{ sec} \quad (70)$$

Thus the time delay until breakdown after the sudden application of a critical field is of the order of nanoseconds.

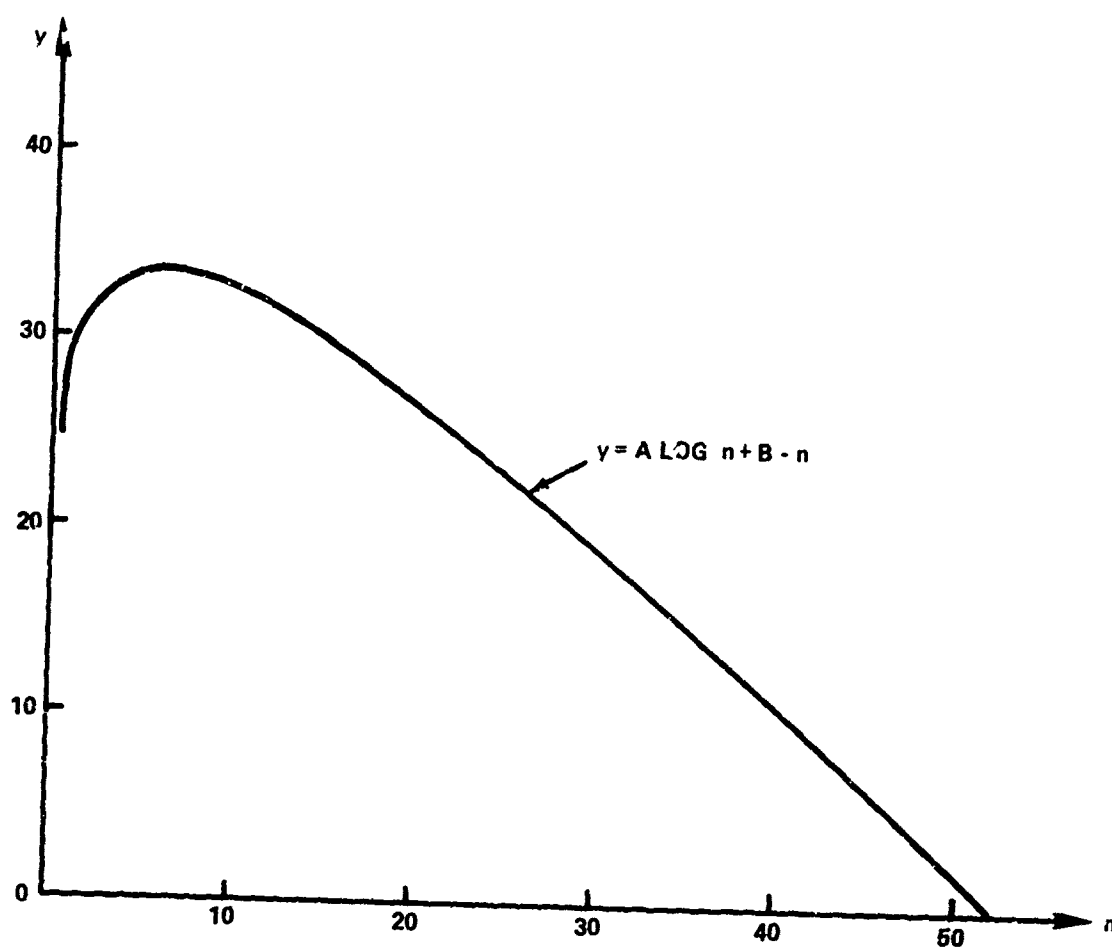


FIGURE 3 GRAPHICAL SOLUTION OF EQUATION (68) FOR THE COLLISION GENERATION NUMBER n
 $A = 5.771$, $B = 29.347$

7. CHARACTERISTIC RESULTS. The prediction of actual breakdown fields from formulas such as (48) and (49) requires knowledge of various material parameters such as the effective mass, dielectric constants, ionization potentials, etc. Unfortunately many or most of these have not been measured for explosive materials. Lack of an adequate experimental data base precludes accurate predictions of breakdown strength in explosives. Some general characteristics of breakdown may, however, be given. These include:

1. It occurs very rapidly. Experimental breakdowns are observed to occur in less than 10^{-8} sec, with some instances as long as 10^{-6} awx.
2. It is the breakdown of an equilibrium condition and therefore occurs at an essentially constant temperature. Breakdown may thus occur at low or room temperatures.
3. In the type of breakdown we have considered, dielectric strength will be independent of sample geometry.

Other modes of breakdown also exist. In unpublished work recently completed at NSWC J. Forbes measured breakdown fields in a number of explosives. In the experimental setup employed, the explosive sample was sandwiched between two electrodes. Breakdown theory as developed in the previous section would not be applicable as we would expect substantial carrier injection from the electrodes¹⁶. This would lead to lower measured critical fields than might be expected in an "electrodeless" geometry.

8. CRITICAL ELECTRON DENSITY. When the density of electrons is large enough, the single electron-phonon interaction picture is no longer realistic and the effect of electron-electron collisions must be included in any calculation.

The critical density for the validity of the one-electron approximation is obtained by considering the net rate of energy transfer to the lattice. As the rates of absorption and emission of a lattice phonon are given by n_q and $(1+n_q)$ respectively, it will take, on the average $2n_q+1$ collisions to transfer a single quantum of energy $h\omega$. The transfer rate may then be approximated by

$$\frac{dU}{dt}_{\text{LATTICE}} = \frac{1}{(2n_q + 1)} \frac{h\omega}{\tau(E)} \quad (71)$$

¹⁶Fowler, R., and Nordheim, L., "Electron Emission in Intense Electric Fields," Proc. Roy. Soc., A119, 173 (1928).

The rate of energy loss due to collision with other electrons is given by¹⁷ Bohm and Pines as

$$\frac{dU}{dt}_{\text{ELECTRON}} = \frac{4\pi ne^4}{\sqrt{2mE}} \quad (72)$$

The critical electron density for a consideration of electron-electron effects is obtained when (71) and (72) are of equal magnitude. This gives the condition

$$n_{\text{CRITICAL}} = \frac{\sqrt{2mE}}{4\pi e^4} \frac{h\omega}{(2n_q + 1)} \frac{1}{\tau(E)} \quad (73)$$

In polar materials, substitution of some representative values gives $n_{\text{CRITICAL}} = 10^{17} (\text{cm})^{-3}$ typical values for non-polar crystals gives $n_{\text{CRITICAL}} = 10^{14-15} (\text{cm})^{-3}$.

Published values¹⁸ for conductivities in explosive materials give values of σ in the range of $10^{-10} - 10^{-8} (\Omega\text{-M})^{-1}$. Associated number densities are obtained from

$$n = \frac{m\sigma}{e^2\tau} \quad (74)$$

Taking the effective mass equal to the electron mass and using a value of τ of 10^{-14} we obtain $n < n_{\text{CRITICAL}}$ and use of the single electron approximation is therefore justified.

Recently G. Hammond (NSWC) has undertaken some conductivity measurements of explosives in the microwave regime. Preliminary results indicate substantially enhanced conductivities at these frequencies. We shall therefore briefly outline the modifications to the single electron treatment given above which enable treatment of the many body electron distribution.

¹⁷ Pines, D., "A Collective Description of Electron Interactions," Phys. Rev. 92,626 (1953).

¹⁸ Federoff and Sheffield, Ed. Encyclopedia of Explosives D1221 (1972).

9. EFFECTS OF THE ELECTRON DISTRIBUTION. When the many electron system is described by a distribution function f defined in such a way that $\frac{2}{(2\pi\hbar)^3} f d^3q d^3p$ describes the number of electrons in volume element d^3a with wave vector $k = \frac{p}{\hbar}$, the current density may no longer be given by Equation (4) but is now defined by

$$J = \frac{e}{m} \int v f d^3k \quad . \quad (75)$$

In general, only the asymmetrical component of f will give rise to a current. Equations such as (6) and (12) representing probabilities for electronic transitions must be augmented by factors such as $f(k)(1-f(k'))$ which represent the probability $f(k)$ that the initial state $|k\rangle$ is occupied and the final state $|k'\rangle$ is unoccupied. Following this prescription, the current density J and the energy transfer rate B may be computed and a breakdown field F_c derived as in section (5) which, however, accounts for the effects of the electron distribution. It has been implicitly assumed here that the electrons and phonons are thermally equilibrated.

In the high density case, the assumption of an electron-phonon system in thermal equilibrium may no longer be valid. With a sufficient density of conduction electrons the electrons may exchange energy among themselves at a much greater rate than with the lattice. The phonons and electrons must then be treated as a two-fluid system with the phonons at temperature T and electrons at temperature T_e . The applied field imparts momentum to the electrons shifting them away from the 0-field equilibrium momentum to some new p . This particular problem has been treated extensively by Frohlich and Paranjape³ (1956) and we shall outline their calculation here. The equilibrium condition is given by the vanishing of the Boltzmann equation

$$\left(\frac{\partial f}{\partial t}\right)_{\text{LATTICE}} + \left(\frac{\partial f}{\partial t}\right)_{\text{FIELD}} + \left(\frac{\partial f}{\partial t}\right)_{\text{ELECTRON}} = 0 \quad . \quad (76)$$

Interelectronic collisions conserve both energy and momentum so that we have

$$\sum_p p \left(\frac{\partial f}{\partial t} \right)_e = 0 \quad \text{and} \quad \sum_p \frac{p^2}{2m} \left(\frac{\partial f}{\partial t} \right)_e = 0 . \quad (77)$$

Equation (55) then gives the two conditions

$$\sum_p p \left\{ \left(\frac{\partial f}{\partial t} \right)_L + \left(\frac{\partial f}{\partial t} \right)_F \right\} = 0 \quad (78)$$

$$\sum_p \frac{p^2}{2m} \left\{ \left(\frac{\partial f}{\partial t} \right)_L + \left(\frac{\partial f}{\partial t} \right)_F \right\} = 0 . \quad (79)$$

The distribution function is assumed to be of the form

$$f(p) = a e^{-(p - p_0)^2 / 2mkT_e} . \quad (80)$$

The two Equations (78) and (79) are then sufficient, in principle, to determine the two unknowns p_0 and T_e . Calculation of p_0 enables computation of a mean conduction density

$$J = \frac{ne p_0}{m} . \quad (81)$$

The rate of energy transfer to the lattice will be given by

$$B(T_e, T) = \int \frac{p^2}{2m} \left(\frac{\partial f}{\partial t} \right)_L d^3p , \quad (82)$$

where the summation in Equation (79) has been converted to an integral and $\left(\frac{\partial f}{\partial t} \right)_{\text{LATTICE}}$ is obtained formally by considering the probabilities of transitions into or out of states of momentum p .

$$\begin{aligned} \frac{\partial f}{\partial t}_L = & \int_q f(p)P^a(p, p+q) - f(p+q)P^e(p+q, p) \\ & + f(p)P^e(p, p-q) - f(p+q)P^a(p-q, p) . \end{aligned} \quad (83)$$

It is found that Equation (82) has a maximum at some T_e . Setting $J \cdot F$ equal to $B(T, T_e^{\text{MAX}})$ then defines a critical breakdown field.

10. APPLICATION TO EXPLOSIVES. The application of electrical breakdown theory to an explosive medium follows closely breakdown theory in inerts. Since electrical breakdown is perceived as the destruction of steady state-equilibrium condition, the most important consideration is the nature of the electron-phonon coupling. Important parameters are the temperature T the phonon spectrum and the various optical constants. These considerations apply equally well to explosives in metastable equilibrium with their environment as of inerts. It is only in a consideration of thermal breakdown that the exothermic kinetics of explosive materials must be accounted for.

III. THERMAL BREAKDOWN

1. THERMAL BREAKDOWN IN INERTS. In thermal breakdown it is assumed that the properties of the lattice may be fully described by the macroscopic temperature dependent conductivity. As an electric field is applied, energy will be deposited through Joule heating. This deposited energy will raise the temperature of the lattice which results in an increase in the conductivity function. The increased conductivity gives rise to an increased current which implies a higher rate of joule heating and so on. If the lattice is unable to dissipate this energy at a rate great enough to overcome the joule heating, temperatures will rise uncontrollably and thermal breakdown will be said to occur. The critical field is then defined by the greatest field for which a steady state solution of the heat transfer equation (Equation (84)) is possible. Formally, the theory of thermal breakdown in inerts consists of finding solutions to the equation

$$c_v \rho \frac{\partial T}{\partial t} = \nabla \cdot (\kappa \nabla T) + \sigma(F, T) F^2(t) . \quad (84)$$

c_v is the specific heat per unit mass, ρ is the density, κ is the thermal conductivity. σF^2 represents the rate of energy deposited through Joule heating. Equation (84) says that the input energy is partly stored as internal energy tending to increase the temperature and partly conducted away.

Many dielectrics have been found to follow a Poole-Frenkel conductivity¹⁹ law given by

$$\sigma(F, T) = \sigma_0(F) e^{-\phi/kt} \quad (85)$$

ϕ represents a work function for electron ionization from a valence band or a trap to the conduction band. σ_0 may in general be a function of the electric field F .

2. THERMAL BREAKDOWN IN REACTIVE MEDIA. With the identification of the Poole-Frenkel conductivity function the theory of thermal breakdown in inerts is seen to be mathematically equivalent to the theory of thermal explosions with the joule heating term σF^2 taking the place of the usual Arrhenius kinetic term in thermal explosion theory. If we now wish to consider thermal dielectric breakdown in explosive materials, Equation (84) must be augmented by an appropriate heat source term to account for the chemical reactivity of the medium. The appropriate equation for thermal breakdown in explosives is now given by

$$c_v \rho \frac{\partial T}{\partial t} = F^2 \sigma_0 e^{-\phi/kT} + \rho Q Z e^{-E_a/kT} + \nabla \cdot (\kappa \nabla T) \quad (86)$$

Equation (86) can equally well be thought of as representing the influence of an applied electric field on thermal explosion solutions. An applied electric field will influence thermal explosion parameters, e.g. induction times to explosion, only insofar as the term $\sigma_0 F^2 \exp(-\phi/kT)$ is not negligible as compared to the Arrhenius kinetic heat generation term. Alternatively, the concept of thermal breakdown of an explosive remains meaningful only so long as the $J \cdot F$ term is not overwhelmed by the Arrhenius term.

¹⁹Frenkel, J., "On Pre-Breakdown Phenomena in Insulators and Electronic Semiconductors," Phys. Rev. 54, 647 (1938).

As is well known, analytic solutions to Equation (86) may not be written down in general because²⁰ of the highly non-linear source terms. A variety of approximate^{21,22} solution schemes for various geometries have been given in the literature. It is also possible to solve (86) in various limiting cases. These include the steady state regime obtained by setting^{23,24} $\frac{\partial T}{\partial t} = 0$ and the "impulse thermal breakdown" obtained by setting $\nabla \cdot (\kappa \nabla T) = 0$. We have chosen to solve Equation (86) numerically retaining all terms except the surface loss terms, assumed to be small for the boundary conditions chosen.

3. CRANK-NICHOLSON METHOD. Equation (86) is an example of a parabolic partial differential equation with a non-linear source term. The Crank-Nicholson²⁵ scheme provides a numerical algorithm for evaluating (86) which is correct to second order in time and stable for any values of increments Δx and Δt . Figure 4 represents the space-time grid for the numerical calculation. At time $t=0$ the solution is given at all spatial points i . The crank-Nicholson scheme involves writing finite difference approximations for the differential operators at points $x_i, t_{n+\frac{1}{2}}$ where $t_{n+\frac{1}{2}}$ represents points halfway between the known level at n and the unknown time level $n+1$.

Equation (86) is solved in a slab geometry illustrated in Figure 5 with boundaries at $x=0$ and $x=x_f$. The divergence term then takes on the form of a second derivative, the thermal conductivity κ assumed constant. The finite difference approximations to the derivatives are given by

$$\frac{\partial T}{\partial t}_{i,n+\frac{1}{2}} \approx \frac{T_{i,n+1} - T_{i,n}}{\Delta t}, \quad (87)$$

²⁰Chambre, P., "On the Solution of the Poisson-Boltzmann Equation," J. Chem. Phys. 20, 1795 (1952).

²¹Zinn, J., and Mader, C., "Thermal Initiation of Explosives," J. App. Phys. 31, 323 (1959).

²²Enig, J., "Approximate Solutions in the Theory of Thermal Explosions for Semi-Infinite Explosives," Proc. Roy. Soc. A305, 205 (1968).

²³Semenov, N., Chemical Kinetics and Chain Reaction, Oxford U. Press (1935).

²⁴Frank-Kamenetskii, Diffusion and Heat Exchange in Chemical Kinetics (trans. N. Thon) Princeton U. Press (1955).

²⁵Von Rosenberg, D. Methods for Numerical Solution of Partial Differential Equations, Elsevier Press (1969).

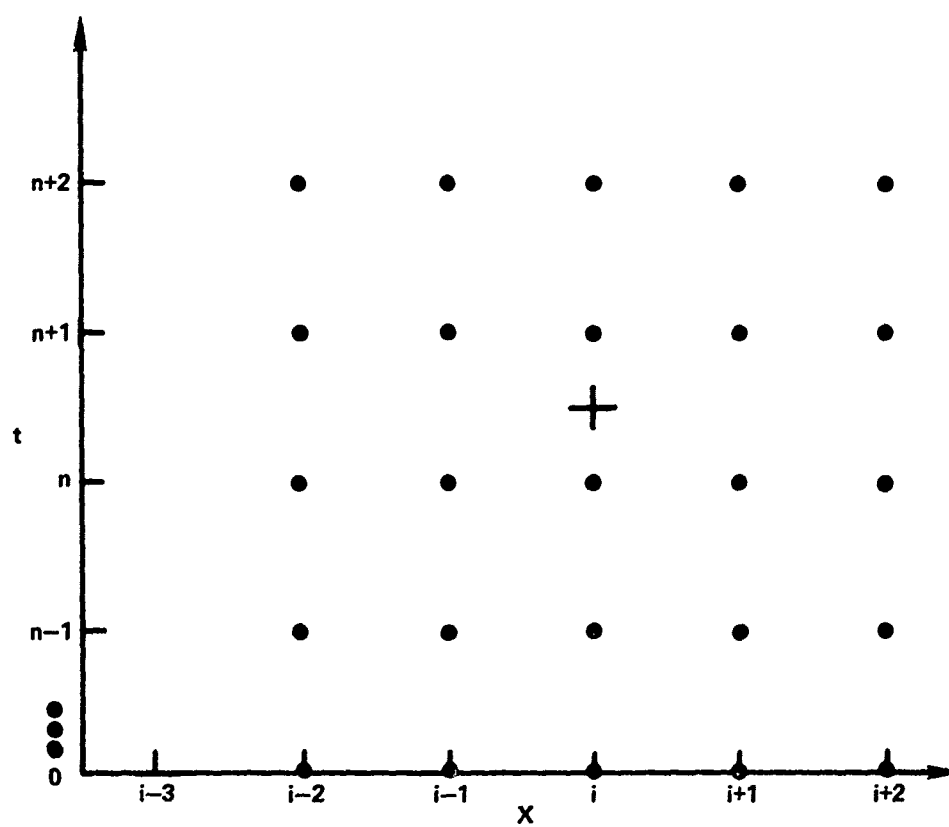


FIGURE 4 GRID FOR FINITE DIFFERENCE COMPUTATION IN A CRANK-NICHOLSON SCHEME

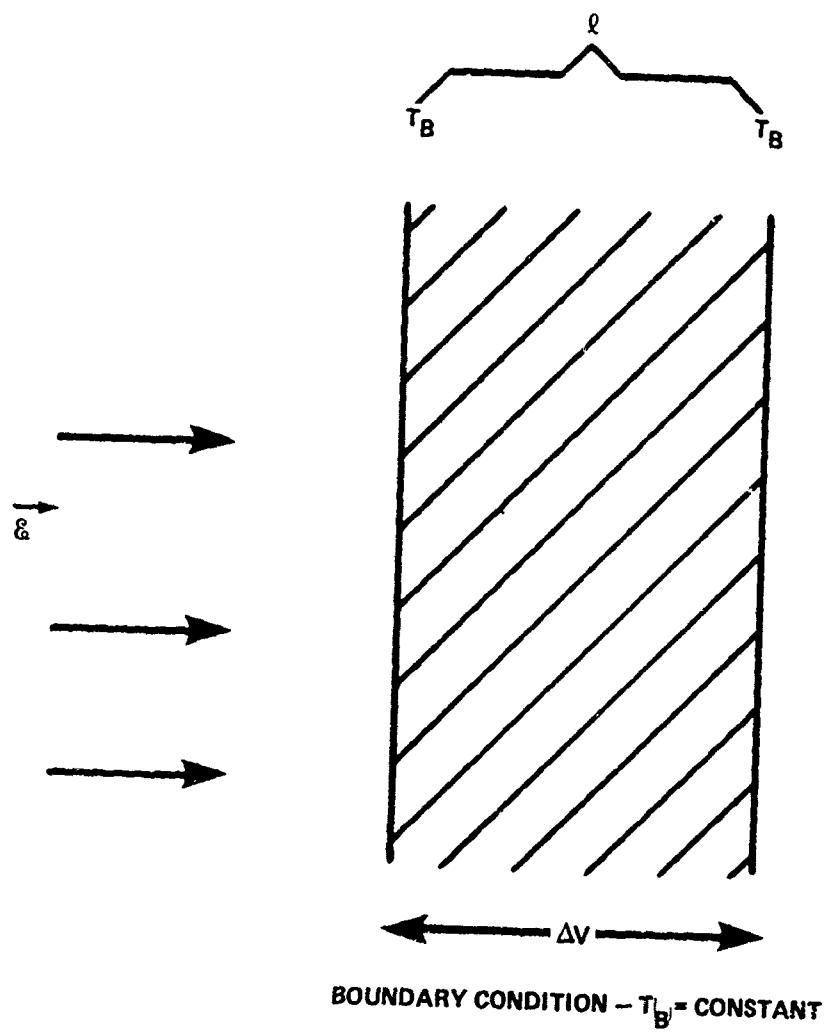


FIGURE 5 COMPUTATIONAL GEOMETRY - A SEMI-INFINITE SLAB

$$\left(\frac{\partial^2 T}{\partial x^2}\right)_{i,n+\frac{1}{2}} = \frac{1}{2} \left[\frac{T_{i+1,n} - 2T_{i,n} + T_{i-1,n}}{(\Delta x)^2} + \frac{T_{i+1,n+1} - 2T_{i,n+1} + T_{i-1,n+1}}{(\Delta x)^2} \right]. \quad (88)$$

The source term is an explicit function of temperature and is estimated by

$$S(T_{i,n+\frac{1}{2}}) = \frac{1}{2} [S(T_{i,n}) + S(T_{i,n+1})]. \quad (89)$$

$S(T_{i,n+1})$ will be given by the approximation

$$S(T_{i,n+1}) = S(T^*) + \left(\frac{\partial S}{\partial T}\right)_{i,n} (T_{i,n+1} - T^*), \quad (90)$$

where

$$T^* = 2T_{i,n} - T_{i,n-1}. \quad (91)$$

The algorithm for the temperatures at the unknown timelevel $n+1$ in terms of known values at time level n is then obtained by substituting (87-91) into (86). We then have

$$\begin{aligned} T_{i-1,n+1} + \left\{ -2 - \frac{2(\Delta x)^2}{\kappa \Delta t} + \frac{\partial S}{\partial T^*} \frac{(\Delta x)^2}{2\kappa} \right\} T_{i,n+1} + T_{i+1,n+1} \\ = -T_{i-1,n} + \left\{ 2 - \frac{2(\Delta x)^2}{\kappa \Delta t} \right\} T_{i,n} - T_{i+1,n} - \frac{1}{2} \left\{ S(T_{i,n}) + S(T^*) \right. \\ \left. - T \left(\frac{\partial S}{\partial T} \right)_{T^*} \right\}. \end{aligned} \quad (92)$$

The boundary conditions are obtained letting the sample sit in a heat bath at temperature T_0 . We then have $T_x = T_{x\text{FINAL}} = T_0$ for all n . Equation (92) defines a set of linear equations in the unknown timestep $n+1$.

Solutions for all the T_i at a given timestep are obtained simultaneously by inverting the coefficient matrix defined by (92). In Appendix B, we display the computer program written for this problem. As is evident from Equation (92) the coefficient matrix is in tridiagonal form. All programs were run on the NSWC/WOL CDC 6500 computer. In Appendix C we explicitly exhibit the tridiagonal matrix and coefficients.

4. DISCUSSION OF RESULTS OF NUMERICAL INTEGRATION. In Figure 6, we show the solutions generated for a typical set of explosive parameters. We have taken $\rho = 1.84 \text{ gm/cm}^3$, $\kappa = 10^{-3} \frac{\text{cal}}{\text{cm} \cdot \text{s}}$, $Q = 1000 \text{ cal/gm}$, $Z = 10^{-14} (\text{sec})^{-1}$, $C_v = .27 \frac{\text{cal}}{\text{gm}^\circ \text{K}}$, and $E_a = 2 \text{ e.v.}$ (1 e.v. $\approx 20,000 \text{ cal/gm}$). We have taken $\sigma_0 = 10^{-7} (\Omega \cdot \text{m})^{-1}$ in line with Picatinney data and have taken the electric field to be a constant during each calculation. The constant chosen ranged from 0 through 10^6 V/cm .

The work function was chosen at 3 e.v. The general results are typical of thermal explosion calculations. The temperature in the sample rises relatively quickly to the bath temperature T_0 or slightly above. If T_0 is chosen below some critical temperature, computed here as 540°K , the temperature stabilizes at T_0 . When T_0 is greater than the critical temperature, the temperature rises initially to T_0 , remains there for some characteristic induction time and then increases very steeply - indicating thermal explosion. There is no indication of electric field dependence in Figure 6 since it was found that for the above choice of parameters the applied electric field has absolutely no effect on the thermal explosion characteristics. It is then clear that the concept of thermal breakdown of an explosive loses validity, being overwhelmed by the chemical production of heat and subsequent thermal explosion at a time well before joule heating alone could raise the temperature significantly.

5. INFLUENCE OF THE CONDUCTIVITY WORK FUNCTION. The critical parameter for possible electric field influence is the work function ϕ . With ϕ chosen as 3 e.v. the joule heating is relatively inefficient even for much larger values of σ_0 corresponding to Hammond's preliminary results. Since there are no available experimental measurements, it seems most reasonable to select ϕ to be somewhat larger than the activation energy E_a . The reason for this is that ϕ represents the energy to ionize an electron into a conduction band while E_a only has to supply enough energy to excite the molecule to the "activated complex" energy state. It is, however, conceivable that under some circumstances ϕ might be less than E_a . This might arise if the conduction mechanism did not arise from a valence to conduction band transition.

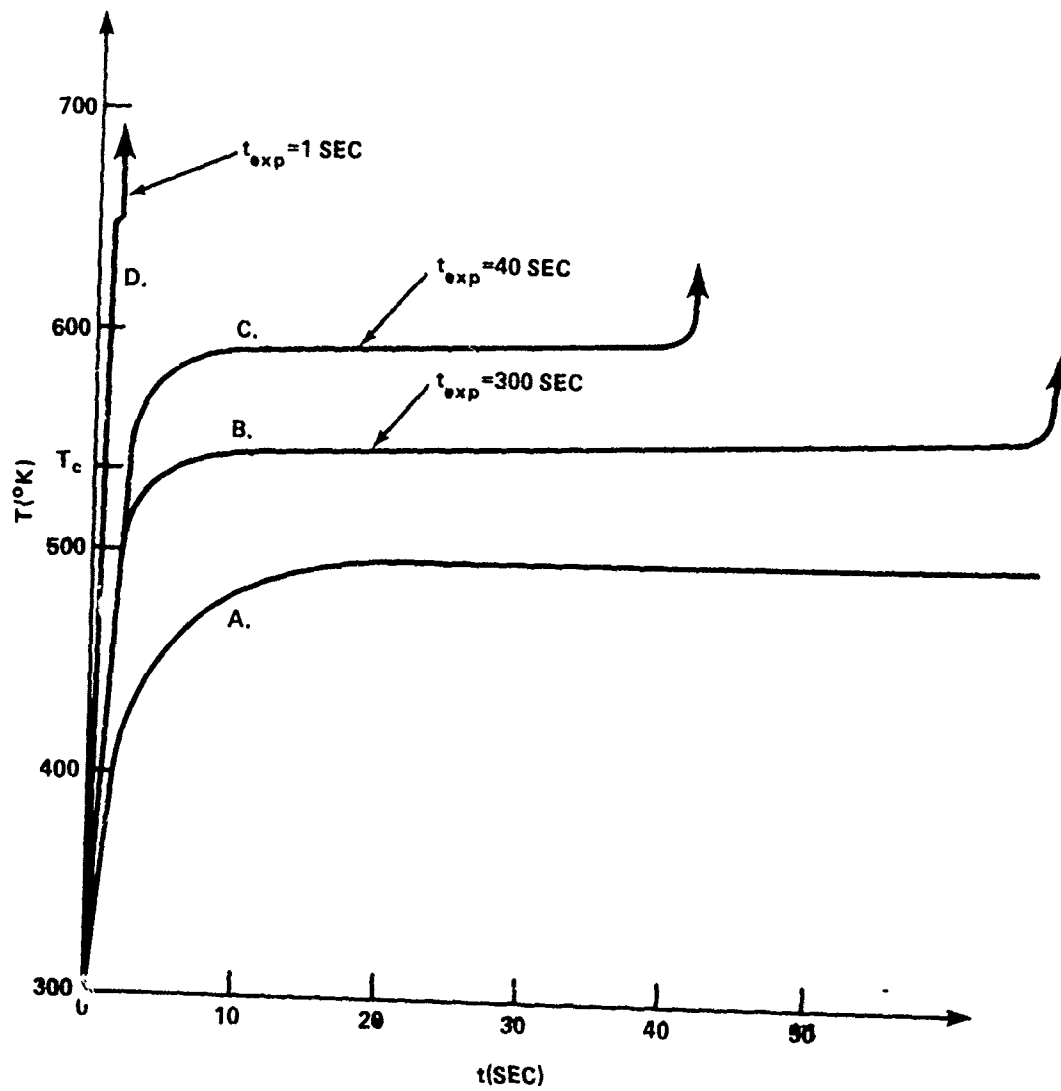
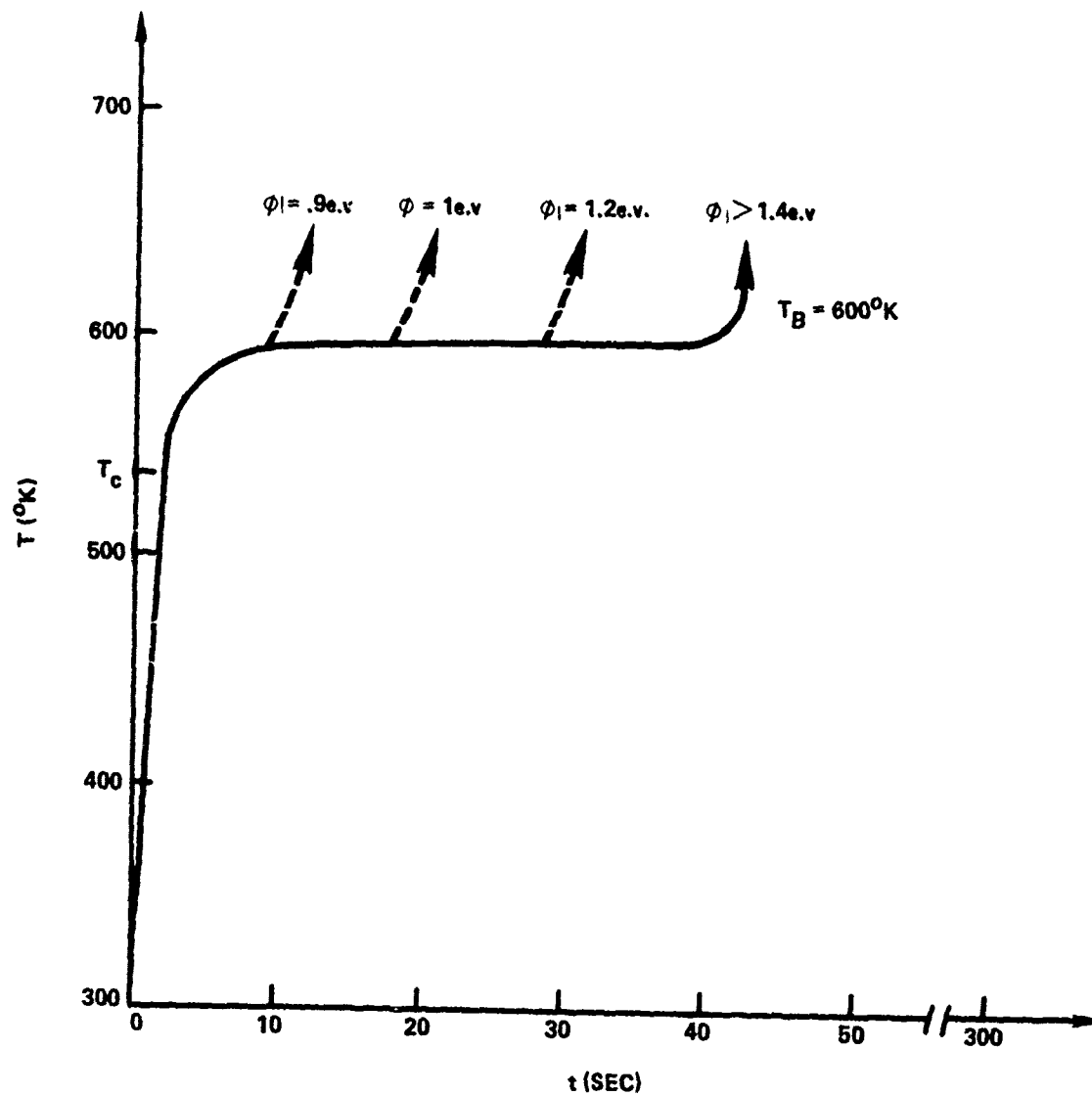


FIGURE 6 TEMPERATURE EVOLUTION FOR DIFFERENT BATH TEMPERATURES T_B .
A. $T_B = 500^{\circ}\text{K}$, B. $T_B = 545^{\circ}\text{K}$, C. $T_B = 600^{\circ}\text{K}$, D. $T_B = 650^{\circ}\text{K}$



PARAMETERS: $\rho = 1.84 \text{ gm/cm}^3$, $z = 10^{14} \text{ SEC}^{-1}$, $Q = 1 \text{ kcal}$, $E_a = 2\text{e.v}$, $C_v = .3 \text{ cal/}^{\circ}\text{K}$,
 $\mathcal{E} = 1.25 \times 10^6 \text{ V/cm}$, $\beta = 10^{-6} [\text{V/cm}]^{-1/2}$, $\sigma_0 = 10^{-7} (\Omega\text{-m})^{-1}$, $\ell = 1 \text{ cm}$.

FIGURE 7 TEMPERATURE EVOLUTION FOR DIFFERENT BATH TEMPERATURE T_B

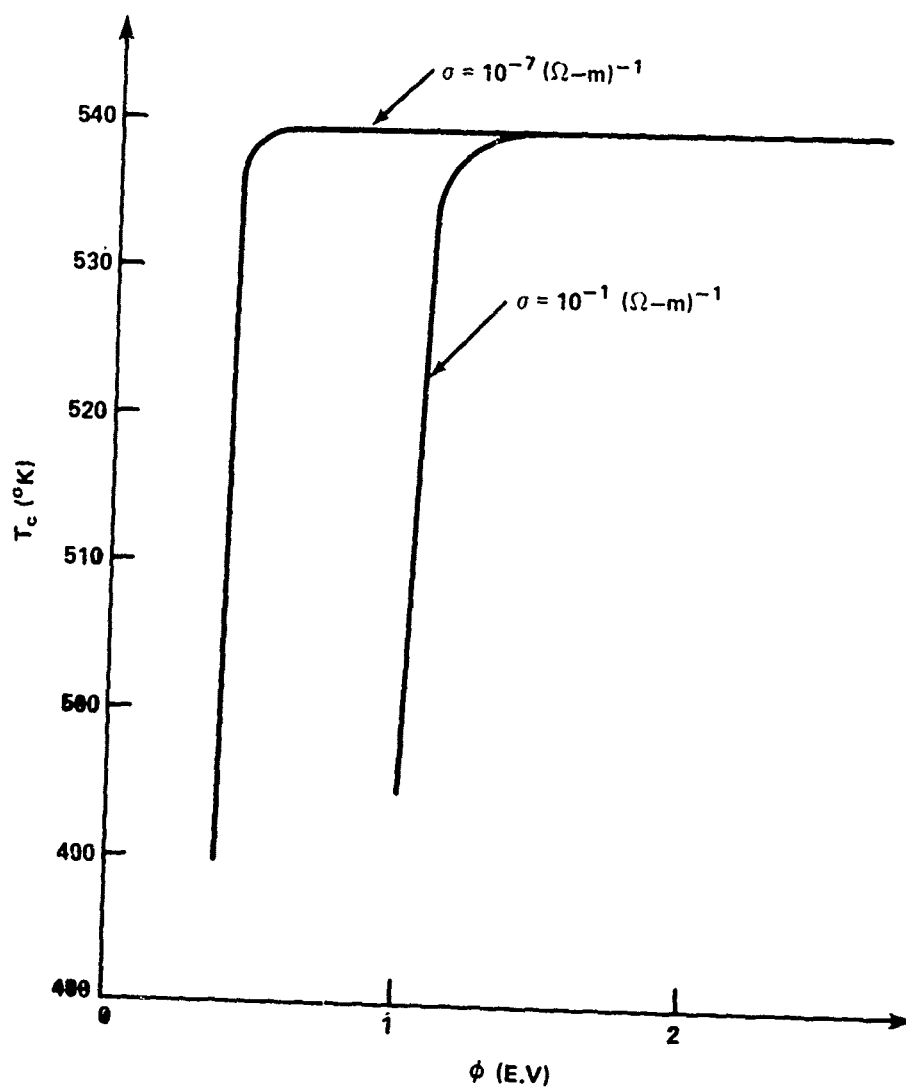


FIGURE 8 DEPENDENCE OF CRITICAL TEMPERATURE FOR THERMAL EXPLOSION ON THE CONDUCTIVITY WORK FUNCTION. ϕ IS IN UNITS OF ELECTRON VOLTS.

One possibility which suggests itself is a conduction due to the ionization of trapped impurities²⁶. The trapping level could lie relatively close to the conduction band and would thus require a smaller work function. In Figures 7 and 8, we have graphed solutions indicating the possibility of radically altering the thermal explosion parameters by applying a strong, step function electric field. The parameters are the same as in Figure 6. The possible tuning of explosion parameters through controlled doping of the explosive sample also suggests itself.

6. THERMAL BREAKDOWN CHARACTERISTICS. The characteristics of thermal breakdown of dielectrics have been seen to be indistinguishable from thermal explosion characteristics. These include

- a. relatively long induction times until explosion
- b. dependence on geometry
- c. high temperature phenomenon

Unlike electrical breakdown which occurs at a constant temperature, thermal breakdown tracks the build up of temperature during the breakdown process.

IV. SUMMARY AND CONCLUSIONS

We have analyzed the application of dielectric breakdown theory to explosive dielectrics. In a lower temperature regime, because the explosive is in metastable equilibrium with its environment, breakdown theory could be modeled after that in inerts. We have predicted electric breakdown fields on the order of 1 MV/cm.

In a higher temperature regime, thermal breakdown theory had to be modified to account for the exothermic kinetics of a reacting medium. It was concluded that the distinction between thermal breakdown and thermal explosion disappears for low enough values of the conductivity work function. At still smaller values of ϕ a possible radical effect of applied electric field on thermal explosion parameters exists.

A resolution of these questions and more accurate predictions of breakdown strengths requires a substantial expansion of our data base on explosive materials. In particular, the absorption spectra of explosive molecules through the infrared is of fundamental importance and should be mapped out. The various material constants such as the effective mass or the high and low frequency dielectric constants could be determined e.g. by Raman or Brillouin scattering experiments. Knowledge of absorption spectra would also be prerequisite to an experimental program designed to measure molecular

²⁶Frohlich, H., "On the Theory of Breakdown in Solid Dielectrics," Proc. Roy. Soc. A188, 521 (1949).

energy relaxation through the newly developed techniques of picosecond spectroscopy²⁷. More measurements of the conductivities of explosive dielectrics are needed. These measurements should be made at various temperatures to test the Poole-Frenkel model and determine activation energies for conduction. Ionization energies for explosive molecules should be measured. To separate out space charge and electron tunneling effects, measurements of electrical breakdown should be made with electrodes at some distance from the explosive surface in an evacuated chamber.

V. ACKNOWLEDGEMENT

I would like to thank Dr. Edward Toton, Dr. Kibong Kim and Dr. Bernard Weinberg for helpful discussions. I would also like to express my appreciation to Jim Coughlin and John Erkman who, wise in the ways of UNITEK, kept our system afloat while the numerical computations were run.

²⁷ Eiseenthal, K., "Picosecond Spectroscopy," Annual Reviews of Physical Chemistry, Vol. 28, 1977.

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APPENDIX A

CALCULATION OF THE INTERACTION CONSTANT

To determine the constant C , we consider the interaction energy of two point charges. Classically this energy is given by

$$E = \frac{e^2}{4\pi r_{1,2} \epsilon} \quad (A-1)$$

where $r_{1,2}$ represents the interparticle distance. The dielectric function ϵ represents the total response of the lattice to the perturbing radiation. This dielectric response is given by

$$\epsilon = \epsilon_{\infty} + \frac{(\epsilon_0 - \epsilon_{\infty})^2 \omega_0^2}{\omega_0^2 - \omega^2} \quad (A-2)$$

ϵ_{∞} represents the high frequency background response due to higher energy electronic resonances. If the polarization due to lattice displacement (i.e., phonon coupling) is ignored the interaction energy would be

$$E = \frac{e^2}{4\pi r_{1,2} \epsilon_{\infty}} \quad (A-3)$$

The net energy due to electron-phonon interaction can therefore be represented by

$$\Delta E = \frac{e}{4\pi r_{1,2}} \left(\frac{1}{\epsilon} - \frac{1}{\epsilon_{\infty}} \right) \quad (A-4)$$

We may also write down the interaction energy shift due to electron phonon interactions from the quantum mechanical perturbation formula

$$\Delta E = \sum_n \frac{|\langle n | H_{e-ph}(r_1) + H_{e-ph}(r_2) | 0 \rangle|^2}{E^{(n)} - E^{(0)}} . \quad (A-5)$$

H_{e-ph} is given by Equation (19) and involves a sum over one-phonon creation and annihilation operators. The matrix element is given explicitly by

$$\langle n | H_{int}(r) | 0 \rangle = \frac{Ce4\pi i}{\sqrt{V}} \sum_q \left[\frac{e^{i\vec{q} \cdot \vec{r}}}{q} a_q - \frac{e^{-i\vec{q} \cdot \vec{r}}}{q} a_q^\dagger \right] | 0 \rangle . \quad (A-6)$$

Applying the selection rules

$$\langle n' | a_q | n \rangle = \sqrt{n+1} \delta_{n+1,n} , \quad (A-7)$$

$$\langle n' | a_q^\dagger | n \rangle = \sqrt{n} \delta_{n-1,n} , \quad (A-8)$$

we have

$$\langle n | H_{int}(r) | 0 \rangle = \frac{4\pi i e C}{\sqrt{V}} \frac{e^{i\vec{q} \cdot \vec{r}}}{q} . \quad (A-9)$$

The denominator of (A-5), $E^{(n)} - E^{(0)}$, represents a single phonon quantum. It has been found that polar optical phonons display little dispersion. The denominator may then be removed from the sum as approximately constant and equal to $\hbar\omega$, with ω the polar optical frequency.

Applying (A-9) in (A-6) and neglecting the self energy term we have for ΔE

$$\Delta E = \frac{-2(r\pi e)^2 C^2}{V\hbar\omega} \sum_q \frac{e^{i\vec{q} \cdot (\vec{r} - \vec{r}')}}{q^2} . \quad (A-10)$$

The summation term is recognizable as a Fourier decomposition of $\frac{1}{4\pi r_{1,2}}$.

We then have

$$\Delta E = \frac{-2C^2 4\pi e^2}{h\omega} \frac{1}{r_{1,2}}. \quad (A-11)$$

Equating (A-10) and (A-11) we the have

$$C = \frac{1}{4\pi} \left(\frac{h\omega}{2}\right)^{\frac{1}{2}} \left(\frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon}\right)^{\frac{1}{2}}. \quad (A-12)$$

APPENDIX B

COMPUTER PROGRAM FOR THERMAL BALANCE EQUATION
UTILIZING A CRANK-NICHOLSON SCHEME

PROGRAM MIKE1 12/75 3-1-1

FIN 4.5+42h

```

PROGRAM MIKE1(INPUT,OUTPUT,TAPE5=OUTPUT)
DIMENSION U(100),B(100),G(100),JP(100),F1(100),F2(100),FS1(100),
*FS2(100),FS(100),US(100),DFS(100),DF(100),B3(100)
DIMENSION F(100)

C
C SET INITIAL PARAMETERS
C
C PARAMETER SET- XL=SLAB THICKNESS,IMAX= NUMBER OF CELLS AT
C LEVEL N,TC=THERMAL CONDUCTIVITY,RO=DENSITY,Z= FREQUENCY
C FACTOR,J=HEAT OF DETONATION,SH=SPECIFIC HEAT,E1=ACTIVATION
C ENERGY(E.V),E2=WORK FUNCTION,SIGMA=CONDUCTIVITY,E=ELECTRIC FIELD.
C
IMAX=100
ILAST=IMAX-1
IQ=IMAX-2
NT=50
DT=.0025
XL=1.
DX=XL/IMAX
TC=1.E-3
RO=1.84
Z=1.E14
G=1.E3
SH=.27
XK=1.
XK=TC/(RO*SH)
E1=2.
E2=3.
SIGMA=1.E-1
DELTA=.01
E=1.25E-1

C
C INITIAL CONDITIONS. BOTH BOUNDARIES OF SLAB ARE AT THE
C BATH TEMPERATURE U0, INTERIOR INITIALLY AT 300 KELVIN.
C U(I) ALWAYS REPRESENTS TEMPERATURES AT THE KNOWN TIME LEVEL N.
C UP(I) ARE THE UNKNOWN TEMPERATURES AT TIME LEVEL N+1. JS(I)
C IS AN INTERPOLATION BETWEEN N AND N+1.
C
U0=555.
U(IMAX)=U0
DO 13 I=1,ILAST
U(I)=300.
13 US(I)=U(I)
C

```

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C BOUNDARY CONDITIONS

```

US(IMAX)=U0
UP(IMAX)=J0
WRITE(5,5)
5  FORMAT(1H1,/,*,* PARAMETER SET*,/)
WRITE(5,12)TC,R0,SH,0,Z,E1,E2,SIGMA,XL,DX,VI,DT,E
12  FORMAT(/1X,*THERMAL CONDUCTIVITY=*,E9.2,* DENSITY=*,F4.2,* SPECIFIC
+HEAT=*,F4.2,/* HEAT OF DETONATION=*,E9.2,* FREQUENCY FACTOR=*,E9.2
+,* ACTIVATION ENERGY=*,E9.2,/* ORK FUNCTION=*,E9.2,* SIGMA=*,E9.2
+,* TOTAL LENGTH(CM)=*,E0.1,/* DELX=*,E9.2,* NO. OF TIMESTEPS=*,I5
+,* DFLT=*,E9.2,/* ELECTRIC FIELD(V/CM)=*,E9.2,///)
E1=E1*1.5022*1.E4/1.38
E2=E2*1.5022*1.E4/1.38
WRITE(5,13)
13  FORMAT(//* INITIAL TEMPERATURE DISTRIBUTION*/)
WRITE(5,22)U0,(U(I),I=1,IMAX)

```

USRAM NIKL: 15/15 CPT=1

IV 4.5+426

18.

```

T=0.
DO 30 V=1,VT
T=T+DT
DELXT=DT/(DX**2)
BP=2./(DELXT*XX)
A1=3*Z*DT*BP/SH
B1=3*SIGMA*(E**2)*DT/(SH*R0)
15  CONTINUE
IF((C.GE.DELTA).OR.(E.GE.DELTA))(GO TO 14)
DO 11 I=1,IMAX
FS(I)=0.
F(I)=0.
DF(I)=0.
DS(I)=0.
BB(I)=-2.-BP
11  CONTINUE
GO TO 17
14  CONTINUE
DO 16 I=1,IMAX
F1(I)=A1*EXP(-E1/J(I))
F2(I)=B1*EXP(-E2/U(I))
F(I)=F1(I)+F2(I)
D=(F1(I)+F2(I)+F2*(I))/(U(I)**2)
FS1(I)=A1*EXP(-E1/JS(I))
FS2(I)=B1*EXP(-E2/JS(I))
FS(I)=FS1(I)+FS2(I)
DFS(I)=(E1*FS1(I)+E2*FS2(I))/(US(I)**2)
GO 16
SET COEFFICIENTS OF TRIAGONAL MATRIX
BB(I)=-2.-BP+DFS(I)/2.
16  CONTINUE
17  CONTINUE
BD=2.-BP
GO 16
INVERT TRIAGONAL MATRIX,SOLVING FOR NEW UP.
G(1)=BB(1)
G(1)=(BD*U(1)-U(2))/BB(1)
G(1)=G(1)-2.*U0/BB(1)
DO 40 I=2,ILAST
B(I)=BB(I)-1./B(I-1)
J=-J(I-1)+BD*U(I)-J(I+1)-(F(I)+FS(I)-JS(I)*D*FS(I))/2.
G(I)=(D-G(I-1))/B(I)
40  CONTINUE
JP(ILAST)=G(ILAST)-JP(IMAX)/B(ILAST)
DO 30 J=1,IS
I=ILAST-J
JP(I)=G(I)-JP(I+1)/B(I)
50  CONTINUE

```

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```

C CHECK CONVERGENCE
  DO 60 I=1,IMAX
    IF (ABS(UP(I)-US(I)).LE.DELTA) GO TO 60
    DO 58 II=1,IMAX
      US(II)=UP(II)
    58 CONTINUE
    GO TO 15
  60 CONTINUE

```

PROGRAM MIKE1 13/73 UPI=1

FIN 4.6+928

```

C
  WRITE(6,20)N,DT,T
  WRITE(6,22)J0,(JP(I),I=1,IMAX)
20 FORMAT(/'N=',I4,' I4,' DT=',F12.5,' ELAPSED TIME=',F12.5)
22 FORMAT(10E12.3)
C
  RESET J(I) FOR NEXT TIME STEP COMPUTATION
  DO 70 I=1,IMAX
    US(I)=2.*JP(I)-U(I)
  70 U(I)=UP(I)
    DT=1.4*DT
  30 CONTINUE
  80 CONTINUE
  STOP
  END

```


APPENDIX C

THE TRIDIAGONAL MATRIX AND THOMAS ALGORITHM

Equation (71) is of the form

$$a_i u_{i-1} + b_i u_i + c_i u_{i+1} = d_i, \quad (C-1)$$

where the u_i represent the unknown temperatures at the spatial points i at time level $n+1$. The coefficients are equal to

$$a_i = c_i = 1, \quad (C-2)$$

$$b_i = -2 - 2 \frac{(\Delta x)^2}{\kappa \Delta t} + \left(\frac{\partial S}{\partial T} \right)_{T^*} \frac{(\Delta x)^2}{2\kappa}, \quad (C-3)$$

$$d_i = T_{i-1,n} + \left\{ 2 - 2 \frac{(\Delta x)^2}{\kappa \Delta t} \right\} T_{i,n} - T_{i+1,n} - \frac{1}{2} \{ S(T_{i,n}) + S(T^*) - T^* \left(\frac{\partial S}{\partial T} \right)_{T^*} \}. \quad (C-4)$$

The tridiagonal matrix equation for each n then takes the form

$$\begin{bmatrix} b_1 & c_1 & 0 & & \\ a_2 & b_2 & c_2 & & \\ 0 & a_3 & b_3 & c_3 & 0 \\ & & & & 0 \\ 0 & & & a_{i_{\max}-1} & b_{i_{\max}-1} \end{bmatrix} \begin{bmatrix} u_1 \\ u_2 \\ \\ u_{i_{\max}-1} \end{bmatrix} = \begin{bmatrix} d_1 \\ d_2 \\ \\ d_{i-1} \\ d_{\max} \end{bmatrix} \quad (C-5)$$

This matrix is inverted in Appendix B using the Thomas algorithm. This involves computing β_i and γ_i such that

$$\beta_1 = b_1, \quad (C-6)$$

$$\beta_i = b_i - \frac{a_i c_{i-1}}{\beta_{i-1}}, \quad (C-7)$$

$$\gamma_1 = \frac{d_1}{b_1}, \quad (C-8)$$

$$\gamma_i = d_i - \frac{a_i \gamma_{i-1}}{\beta_i}. \quad (C-9)$$

We then have for the unknown u_i

$$u_{i_{\max}-1} = \gamma_{i_{\max}-1}, \quad (C-10)$$

$$u_i = \gamma_i - \frac{c_i u_{i+1}}{\beta_i}. \quad (C-11)$$

It should be remembered that $u_{i_{\max}}$ is known from the boundary conditions.

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